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The phonon Bragg switch: a proposal to generate sub-picosecond X-ray pulses

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Abstract

We discuss a method to modulate an X-ray beam that relies on the scattering of hard X-ray photons by a superlattice of optical phonons generated by ultrafast laser pulses incident on a Bragg crystal. Using commercially available laser systems and well-developed techniques for pulse shaping, the crystal lattice motion can be controlled so that efficient Bragg diffraction is switched on and off in a time comparable to a phonon oscillation period (typically, 0.1–1 ps). A detailed proposal is given for the case of GaAs. © 1999 Elsevier Science Ltd. All rights reserved.

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Third-generation synchrotrons are bright sources of tunable X-rays for applications in physics, chemistry, biology, and engineering. Most spectroscopic or imaging problems make use of the high average spectral brightness of these sources, which can exceed 10^{12} X-rays per second within a 10^{-4} fractional energy band, with beam divergence below 100 μr. Still more impressive is the peak spectral brightness, which can approach 10^4 monoenergetic hard X-rays per picosecond. This is high enough for diffraction or absorption measurements on the time scale of atomic motion in molecules and condensed media.

The X-ray pulse duration from a synchrotron is on the order of 100 ps, which is too long to resolve motion on the 0.1–1 ps scale of molecular dynamics, so ultrafast X-ray gates or X-ray detectors are needed. Here we propose a new method to modulate a synchrotron X-ray beam that relies on the scattering of hard X-ray photons by coherent optical phonons generated by ultrafast laser pulses incident on a Bragg mirror. This transient mirror can be configured as an X-ray slicer, to switch sub-picosecond bursts of X-rays out of a single bunch delivered by the synchrotron. The reflectivity can be close to unity during the time when the mirror is gated "on". Further, the X-ray switch duration can be controlled, and the switch can be turned on and off several times during a single pulse of the synchrotron.

Recent attempts to produce ultrashort pulses of X-rays have included short bursts of K_α radiation from laser-induced plasmas [1,2], and laser-induced ponderomotive scattering of the electrons in a synchrotron storage ring [3–5]. Still another approach uses a short laser pulse to melt a Bragg crystal and interrupt X-ray diffraction [6]. These X-ray experiments corroborate earlier light scattering conclusions that the translational order is lost at least as fast, and possibly much faster, than the onset of melting [7–11].

We propose to excite a Bragg mirror coherently, so

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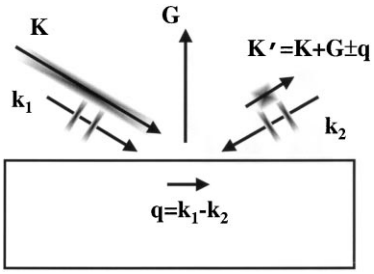

Fig. 1. Geometry for the Bragg switch. Optical pulse trains with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ create and terminate a transient standing wave phonon lattice with periodicity $2\pi/|\mathbf{q}|$. An ultrafast slice of the X-rays with incident wave vector $\mathbf{K}$ can Bragg scatter from the superlattice if it satisfies the Bragg condition $\Delta\mathbf{K} = \mathbf{G} \pm \mathbf{q}$, where $\mathbf{G}$ is a reciprocal lattice vector in the crystal.

that instead of melting, the mirror acquires an additional periodic structure or superlattice. The problem divides itself into two parts. First, we show that coherent lattice distortions can be introduced rapidly and reversibly in crystals used for X-ray Bragg mirrors. Second, we discuss how distortions can be introduced with high enough superlattice periodicities (i.e. large wavevectors) to significantly alter the Bragg angle.

Several experiments have shown recently that optical excitation of a crystal can lead to large high-frequency periodic distortions of the lattice [12,13]. To date this effect has only been observed indirectly, through the time-periodic change of the optical constants as the material oscillates at the frequencies of the optical phonons. Relevant to our proposal are the small oscillations induced in GaAs following weak excitation by ultrafast optical pulses above the direct gap [14]. Much larger amplitudes, up to several percent of the natural bond length, have been seen in various materials following intense photoexcitation [15]. These results have been analyzed in terms of a model for which the lattice behavior is similar to that of the vibrational wave packet that forms following resonant electronic excitation of a molecule.

Excitation below the band gap leads to lattice motion without electronic excitation. Here, the mechanism by which light couples to the vibrations is impulsive stimulated Raman scattering [12,13], and the relative amplitude of oscillations is comparable to those measured in opaque materials [16]. The fact that we can excite the lattice and not the electrons has several advantages for our application. If the material remains in the ground electronic state, there is very little incoherently absorbed energy that could lead to heating and melting. Also, large volumes can be excited uniformly in transparent materials. This is advantageous because the optical absorption depth for excitation above the band gap is, in most cases, much smaller than the X-ray scattering length.

Large amplitude optical phonon distortions do not, by themselves, change the Bragg condition. Although they lead to a rearrangement of the atomic positions in a unit cell, the distortions do not change the cells' size and thus, the gross periodicity is unchanged. To produce a new Bragg angle that is not present naturally in the material, we need to organize the pattern of ion distortions into a superlattice, i.e. we need to excite vibrations with high wavevectors, and the higher the better. The highest wavevector that can be produced in a Raman process is attained in the backscattering geometry. In this configuration, two laser beams with photon momentum $\hbar(2\pi n/\lambda)$ impinge the crystal from opposite directions (n is the refractive index), and the lattice scatters photons from one beam into the other one, acquiring momentum $\hbar q \simeq \hbar(4\pi n/\lambda)$ in the process. This results in a superlattice of periodic amplitude distortions interspersed with nodes, with a period of $2\pi/q = \lambda/(2n)$. For various reasons, the backscattering geometry is not very convenient for our application. As shown in Fig. 1, for the case of X-ray scattering from the (001) surface of GaAs, the preferred geometry involves two beams incident at angle θ_i. For this case, the superlattice period is $\lambda/(2 \sin \theta_i)$ which is larger than that for backscattering.

The Hamiltonian describing coupling of the laser field to the phonon is [7]:

$$H = Q \sum_{i,k} (R_{ik} E_i E_k)/2,$$

where $Q(r, t)$ is the amplitude of the phonon field that measures the lattice distortion, R_{ik} the nonlinear Raman susceptibility tensor and $\mathbf{E}$ is the electric field. This interaction leads to a force density acting on the ions, given by $\partial H/\partial Q$. Coherent phonons can be generated using one or more pump pulses. In the single-pump configuration, however, the phonon wavevector is $\approx n\Omega/c$ [7] which is too small for X-ray applications; Ω is the phonon frequency. For the two-pump case, we write the components of the

electric field associated with the pulses *inside* the crystal as $E_m^s = \mathscr{E}_s(u_s)\sin(\omega u_s + \phi)\cos(\alpha_m^s)$, where $s = 1, 2$ and $u_s = t - \mathbf{k}_s\cdot\mathbf{r}/\omega$. $\mathscr{E}_s$ is the slowly varying amplitude and α_m^s are the direction cosines of **E**. Assuming that $|k_1| = |k_2| = n\omega/c$ and that the two pulses have the same amplitude, the relevant driving term, i.e. the crossed term, is

$$F(\mathbf{r},t) = \frac{R\mathscr{E}^2(u)}{4}\cos(\mathbf{q}\cdot\mathbf{r}),$$

where $R = \sum_{ik} R_{ik}\cos(\alpha_i^1)\cos(\alpha_k^2)$, $\mathbf{q} = (\mathbf{k}_1 - \mathbf{k}_2)$ and $u = r_\perp c$) where $r_\perp$ is the component of **r** perpendicular to **q**. Note that in the case where the two beams enter the solid at grazing angle, $|\mathbf{q}| \approx 4\pi/\lambda$. If we ignore phonon dispersion, the equation of motion for the lattice field is $\mathrm{d}^2Q/\mathrm{d}t^2 + \Omega^2$, $Q = \mathrm{F}(\mathbf{r},t)$. For pulses of width $\ll \Omega^{-1}$ (impulsive limit), the solution is

$$Q(\mathbf{r},t) = \int_{-\infty}^{t} \frac{\sin[\Omega(u-\tau)]}{\Omega} F(\mathbf{r},\tau)\,\mathrm{d}\tau$$

$$\approx Q_0\Theta(u)\sin(\Omega u)\cos(\mathbf{q}\cdot\mathbf{r}). \qquad (1)$$

Here Θ is the step function and $Q_0 = R(4\Omega)^{-1}\times \int \mathscr{E}^2(\tau)\,\mathrm{d}\tau$. Thus we obtain the well-known result that the magnitude of the lattice distortion is linearly proportional to the total laser pulse energy. In the problem of interest, the effective width of the phonon superlattice (the X-ray extinction or absorption depth), l, is such that $nl/c \ll 2\pi/\Omega$. As a result, in Eq. (1) we can set $r_\perp \approx 0$.

The optical phonon superlattice created impulsively in GaAs is expected to persist for up to tens of picoseconds [14] as the optical phonon decays due to coupling to acoustic modes. While decay provides a natural termination for the superlattice, faster switching results if we turn off the oscillation coherently by applying a second laser pulse (or pulses) after a half-integer number of optical phonon periods. As demonstrated in the recent coherent phonon experiments in semimetals [17], this can be achieved by dividing the pulses into two parts using a beam splitter and an optical delay line, so that some of the energy arrives in a second set of pulses at the appropriate time delay to de-excite the crystal. With this, the width of the X-ray switch can be controlled from half of a phonon period (about 100 fs in GaAs) to ten picoseconds or more. Materials such as GaP or diamond have higher optical phonon frequencies, so that this technique could be extended to produce switches with less that 20 fs duration.

Consider now the scattering of X-rays by the lattice in the presence of the coherent-phonon superlattice. In general, the scattering intensity for wavevector transfer $\Delta\mathbf{K}$ is

$$\mathscr{I}(\Delta\mathbf{K}) = \left|\sum_{\mathbf{n},j} \mathrm{e}^{\mathrm{i}\Delta\mathbf{K}\cdot\mathbf{r}_{\mathbf{n},j}}\right|^2,$$

where the sum is over all the atoms in the crystal (the indices **n** and j label, respectively, the unit cells and the atoms in a single cell). Let us write $\mathbf{r}_{\mathbf{n},j} = \mathbf{r}_{\mathbf{n},j}^{(0)} + \Delta\mathbf{r}_{\mathbf{n},j}$, where $\Delta r_{\mathbf{n},j}$ is the displacement from equilibrium. To lowest order in Q, and for $nl/c \ll 2\pi/\Omega$, we have

$$\mathscr{I}(\Delta\mathbf{K}) = \left|\sum_{\mathbf{n},j} \mathrm{e}^{\mathrm{i}\Delta\mathbf{K}\cdot\mathbf{r}_{\mathbf{n},j}^{(0)}}(1 + \mathrm{i}\Delta\mathbf{K}\cdot\Delta\mathbf{r}_{\mathbf{n},j})\right|^2 \qquad (2)$$

$$= N^2\Bigg|\delta\Delta\mathbf{K},\mathbf{G}\sum_j \mathrm{e}^{\mathrm{i}\Delta\mathbf{K}\cdot\mathbf{r}_{0,j}^{(0)}}$$

$$+\mathrm{i}\delta_{\Delta\mathbf{K}\pm\mathbf{q},\mathbf{G}}\sum_j\left[\frac{(\Delta\mathbf{K}\cdot\mathbf{e}_{0,j})}{\sqrt{V_\mathrm{C}M_j}}\mathrm{e}^{\mathrm{i}\Delta\mathbf{K}\cdot\mathbf{r}_{0,j}^{(0)}}\right]$$

$$\times Q_0\sin(\Omega t)\Theta(t)/2\Bigg|^2,$$

where M_j and $\boldsymbol{e}_{0,j}$ are the mass and the unit polarization vectors for the jth-ion, and V_C is the volume of the unit cell. This expression is the main result of our work. Contained inside the absolute value brackets is the scattering amplitude as a function of the scattered wave vector $\Delta\mathbf{K}$, the reciprocal lattice vector for the scattering **G**, and the phonon superlattice wavevector **q**. The coherent phonon leads to a new series of Bragg peaks at angles give by the phase-matching condition $\Delta\mathbf{K} = \mathbf{G} \pm \mathbf{q}$. If the crystal is oriented slightly off the main Bragg reflection and in the proper direction, a light-induced diffraction can occur whose temporal duration can be controlled to be a half-integer number of optical phonon periods. The new Bragg reflections are displaced from those at $\Delta\mathbf{K} = \mathbf{G}$ by an angle on the order of $|\mathbf{q}/\mathbf{G}| \approx 5\times10^{-4}$. Relative to a fully allowed Bragg scattering amplitude, the

amplitude A_P of the phonon-induced peaks is suppressed relative to the main Bragg amplitude A_0 by the factor $A_P/A_0 \simeq \mathbf{G}\cdot(\sum_j \Delta\mathbf{r}_{0,j})$ Let I be the integrated intensity of a pulse. From the available data [12], we can infer values of $|\Delta\mathbf{r}|/I$ in the range 10^{-3}–5×10^{-2} Å J^{-1}/cm^2 [18]. For strong excitation, this gives values of A_P/A_0 as large as 10^{-2}–10^{-1}. It is important to note that the sum in Eq. (2), over the index j of the atoms within one unit cell, differs from the conventional expression for the structure factor and that, as a result, forbidden diffractions can become allowed. For example, in silicon, the structure factor for (002) is zero because of destructive interference between the two identical atoms in the cell. However, the phonon-induced Bragg peaks are allowed because the signs of $\boldsymbol{e}_{0,j}$ are opposite for the two atoms (here, we use the standard notation of (*nlm*) to represent the Miller indices of the diffraction wave vector **G**).

In the following, we estimate the performance of the X-ray switch for the specific case of a GaAs Bragg reflector for 10 keV X-rays. To obtain quantitative predictions for the Bragg angle and the scattering efficiency, we consider the geometry shown in Fig. 1. Let us first focus the attention on the geometry for coherent phonon generation. The two laser beams $\mathbf{E}_1 = \hat{y}\mathscr{E}_0 \sin(\mathbf{k}_1'\cdot\mathbf{r} - \omega t)$ and $\mathbf{E}_2 = (\hat{z}\cos\theta_i + \hat{x}\sin\theta_i)\mathscr{E}_0 \sin(\mathbf{k}_2'\cdot\mathbf{r} + \omega t)$ are incident on a (001) crystal surface from nearly opposite directions. Here $\mathbf{k}_1' = k'(\hat{z}\cos\theta_i + \hat{x}\sin\theta_i)$ and $\mathbf{k}_2' = k'(\hat{z}\cos\theta_i - \hat{x}\sin\theta_i)$ where k' is the magnitude of the optical wavevector *outside* the crystal. This geometry was chosen primarily because (i) it gives phonon wavevectors that are significantly larger than those for one-pump configurations and (ii) it allows us to couple, through R_{xy}, to the optical mode of GaAs, of symmetry $\Gamma_{15}(T_2)$. Note that, because their polarizations are crossed, this "linear-perpendicular-linear" geometry does not produce a periodic variation of the intensity at the surface. Instead, there is a periodic linear to circular polarization variation along $\hat{x}$, with period $2\pi/q = 2\pi/(k'\sin\theta_i)$. The continuity of the parallel component of the field at the crystal surface requires that this period is the same *inside* the crystal, where the field excites the optical phonons. The coupling is with transverse, as opposed to longitudinal modes, because R_{xy} involves ion displacements along $\hat{z}$, which is nearly parallel to $\Delta\mathbf{K}$.

X-rays which satisfy the ordinary Bragg condition $\Delta\mathbf{K} = \mathbf{G}$ for an allowed diffraction peak such as (004) in GaAs have an extinction depth that is much smaller than the incoherent attenuation length. For example, just below the absorption edge in GaAs, near 10 keV, the extinction depth is only about 2 μm, while the attenuation length is ≈50 μm. This rapid extinction means that we only need to consider the overlap of the laser beams and the X-rays in the near-surface region. The maximum phonon amplitude that can be excited is limited by multiphoton-induced damage. For light below the GaAs band edge, at ≈1.5 eV, irreversible damage occurs for intensities on the order of 10^{12} W cm^2. From the coherent-phonon data for GaAs involving 2 eV photons [14], we estimate $|\Delta\mathbf{r}|/I \approx 5\times10^{-3}$ Å J^{-1}/cm^2 which is consistent with measurements of the spontaneous Raman cross section [19]. If we use the value of the cross section at the band gap resonance [19], we obtain instead $|\Delta\mathbf{r}|/I \approx 1.5\times10^{-2}$ Å J^{-1}/cm^2. At the damage threshold and for, say, 50 fs pulses, we get at the (004) peak $A_P/A_0 \approx 0.2$. As a result the extinction depth at this angle will increase. The amount of the increase depends on the coherence of the beam. Another way of stating this is that the Darwin width for the superlattice diffraction peak could be a factor of ten narrower than the width of the (004) peak. In practice, imperfections in the crystal and other problems such as heat-induced stress might limit the reflectivity at the superlattice Bragg angle, but we estimate that the reflectivity should be at least 1%, and could be close to 100% as it is for the main peak. The displacement angle of a half-milliradian from the main peak should make this new reflection readily visible above the coherent and incoherent backgrounds.

The time structure of the X-ray diffraction will also depend on geometrical factors in the overlap of the X-ray and optical beams. To minimize temporal spreading of the pulse, a simple geometry should be used in which the X-rays co-propagate with one of the two optical beams. This places the optical beams at the X-ray Bragg angle. That will, in turn, limit the area of overlap in the near-surface region to a stripe with length equal to the optical beam diameter, and width formed by the convolution of the two beams as they cross, which is under 100 μm. This area could be increased by lengthening the duration of one of the two laser pulses. As the pulses must overlap in order

to excite the superlattice, lengthening one beam does not make the diffracted X-ray pulse longer, but does increase the effective area of the Bragg mirror.

In conclusion, we have discussed a new type of X-ray modulator, in which a Bragg reflector is excited by an optical phonon superlattice. X-ray scattering from the superlattice can be modulated optically, resulting in the possibility of ultrafast X-ray switches and other types of X-ray modulation. We acknowledge many useful discussions with Roger Falcone and Roy Clarke. This work was supported by the National Science Foundation, Grant No. 9414335 and by the US Army Research Office under Contract No. DAAH04-96-1-0183.

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